



TITLE:

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CITATION:

Yong, Raymond N.. Sustainability of remediated sediments. SANSAL : An Environmental Journal for the Global Community 2007, 2: 1-29

ISSUE DATE:

2007-03

URL:

<http://hdl.handle.net/2433/108254>

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Sustainability of remediated sediments

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Abstract: This paper presents a discussion of the long term condition of remediated sediments from a geoenvironmental perspective. It examines the problem of long term maintenance of the remediated condition of treated sediments and establishes a set of goals and requirements for their sustainability. First, a number of basic interacting factors are considered in relation to treatment requirements for minimizing the potential for resuspension and remobilization. Next, a geoenvironmental engineering perspective is used to argue that the most viable means for sustainable remediated sediments preservation is to replace removed contaminated sediments with sediment material that has high attenuation capabilities and augment the natural recovery processes in the sediments. It is recognized that ultimate sustainability requires not only preservation of the remediated condition of sediments, but also habitat restoration and regeneration of biodiversity — two important issues beyond the purview of this paper.

Keywords: sustainability, contaminated sediments, natural recovery processes, remediation

1. Introduction

Lacustrine, fluvial and coastal (marine) sediments receive contaminants from various point and non-point sources. A significant proportion of these are land-based industries associated with agriculture, food processing, manufacturing and energy production. These industries, together with urban centres, municipalities and service industries, such as hospitals and military services, broadcast contaminants — defined in this discussion to include both health-threatening and non health-threatening substances — as noxious airborne particulates, surficial discharge and surface flow, and groundwater-transported noxious and hazardous (toxic) contaminants.

The processes and activities contributing to contamination of the surface sediment layer are shown in pictorial form in Figure 1. The surface sediment layer shown in the figure is contaminated by the pollutants and contaminants

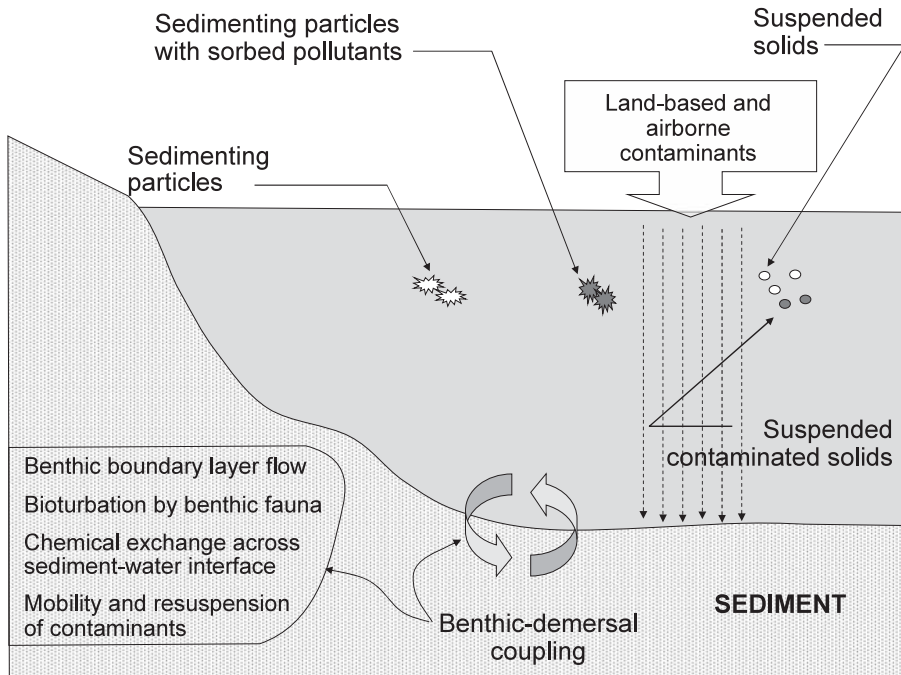


Fig. 1 Processes and activities contributing to contamination of the surface sediment layer

adsorbed (sorbed) onto the sedimenting particles. Note that since some of the airborne particles (solids) introduced to the water environment are very fine, they have the ability to remain suspended in the water environment. Some of these will be contaminated while others may not be. The process of benthic-demersal coupling shown in the figure illustrates the intimate interactions between the benthic layer and the immediate water layer (demersal zone) above the benthic zone. These interactions include; (a) benthic boundary layer flow, (b) bioturbation by benthic fauna, (c) chemical exchange across the sediment-water interface, and (d) mobility and resuspension of sediments.

Contaminated sediments containing large proportions of pollutants impact adversely on biodiversity in the benthic ecosystem. The pollutants (i.e. contaminants that pose a health threat) provide a toxic habitat for demersal or bottom-dwelling fishes and other benthic organisms, many of which are highly tolerant to high concentrations of pollutants accumulating in their tissues. These fish pose serious health threats to humans, through bioaccumulation and biomagnification in the food web. The US Environmental Protection Agency (EPA) estimated in 1998 that 10 per cent of the sediments underlying the surface waters in the US were so contaminated with toxic pollutants that they posed a risk to fish and to humans and wildlife who eat fish (US EPA 1998). Pollutants removed from the contaminated sediments in the Great Lakes Basin included

polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), and heavy metals, for example.

Whereas a notable aim of remediation of contaminated sediments is the removal of opportunities for bioavailability, bioaccumulation and biomagnification of contaminants in the food chain through the removal of the contaminants from the sediments, not all remediation methods currently in use are oriented directly towards this aim. Geoenvironmental engineering strategies for the remediation of contaminated sediments have historically focused on technologies that would ultimately “clean up” or decontaminate the sediments. By and large, the decision-making process determining the technology chosen takes account of a number of factors, including (i) the dynamic nature of sediments, (ii) the highly variable state of the contaminated sediments, i.e. the degree and extent of contamination, (iii) the immense volume of contaminated sediments — conservatively estimated in the millions of cubic metres, and (iv) the level of decontamination desired. Of these, items (i) and (ii) reflect the highly variable nature of the various forces associated with river and tidal currents, sediment deposition sources and rates, and pollutant sources and manner of delivery to the sediments. These factors are not only site and situation specific, they are also time dependent (the time dependency factor in this case referring to both elapsed time and seasonal time).

More recently, it has been recognized that another step in the remediation process is necessary — beyond the obvious decontamination process required to “clean-up” sediments. Returning a remediated sediment to its original uncontaminated state, whatever that might be, is now seen to be a prime requirement of a successful remediation process. This next step is by no means easy: it requires additional knowledge and expertise beyond the range commonly needed in the engineering techniques used to decontaminate contaminated sediments. It would appear necessary not only to establish a benchmark that defines the “uncontaminated natural state”, but also the means and technologies to ensure that a remediated sediment returns to its natural state, replete with its original benthic ecosystem. A detailed discussion of this next step is vital and offers exciting directions for future research beyond the scope of the current paper.

2. Remediated sediments: a problem definition

Remediated sediments are generally considered by most practitioners in the field of contaminated sediments remediation to mean contaminated sediments that have been treated in one fashion or another, to remove or immobilize contaminants in the sediments, with the ultimate aim of eliminating their

bioavailability (bioavailability refers to the degree to which a contaminant or pollutant in a potential source is free for uptake, i. e. movement into or onto an organism). This recognizes the fact that the return of a remediated sediment to its uncontaminated natural state is not a realistic goal, for the reasons discussed above. It is reasoned, with some justification, that if a given remediation procedure can effectively remove the threat of bioavailability, the associated threats to human health of bioaccumulation (uptake and storage of a pollutant by an organism in the food chain), bioavailability and biomagnification (the accumulation of bioaccumulated pollutants in the lower trophic levels to the higher trophic levels) will be eliminated. This prompts three obvious choices of procedure: (1) the complete removal of contaminants, (2) complete detoxification of the toxic contaminants and neutralization of the hazardous contaminants, and (3) total immobilization of the toxic and hazardous contaminants to render them unavailable for interaction with benthic biota. In each case, the denial of the bioavailability of contaminants by removing the opportunities for bioaccumulation and biomagnification is considered the primary objective.

2.1 Remediation, habitat restoration and regeneration of biodiversity

More recently, as aforementioned, a second objective has started to influence the choice of strategies relating to contaminated sediment remediation: that is, the restoration of habitat and regeneration of biodiversity. As is evident from Figure 2, this objective depends first upon the successful denial of bioavailability of contaminants made possible through conventional remediation technology. To date, insufficient attention has been paid to habitat restoration and regeneration of biodiversity in terms of the technology developed and applied in remediating sediments, conceivably because the significant health threats posed by many contaminated sediments in various locations have propelled technology development efforts in the direction of threat reduction. A review of the remediation technologies available would show that most, if not all, remediation technologies address only the first, and not the second, of the two possible objectives of contaminated sediment remediation, with the second yet to be directly or fully addressed.

2.2 Sustainability of remediated sediments

Figure 2 summarizes the above discussion by raising the following question: what is the end point for the remediation of contaminated sediments — the elimination of bioavailability of contaminants only, or the elimination of bioavailability and restoration of habitat and regeneration of biodiversity? Two

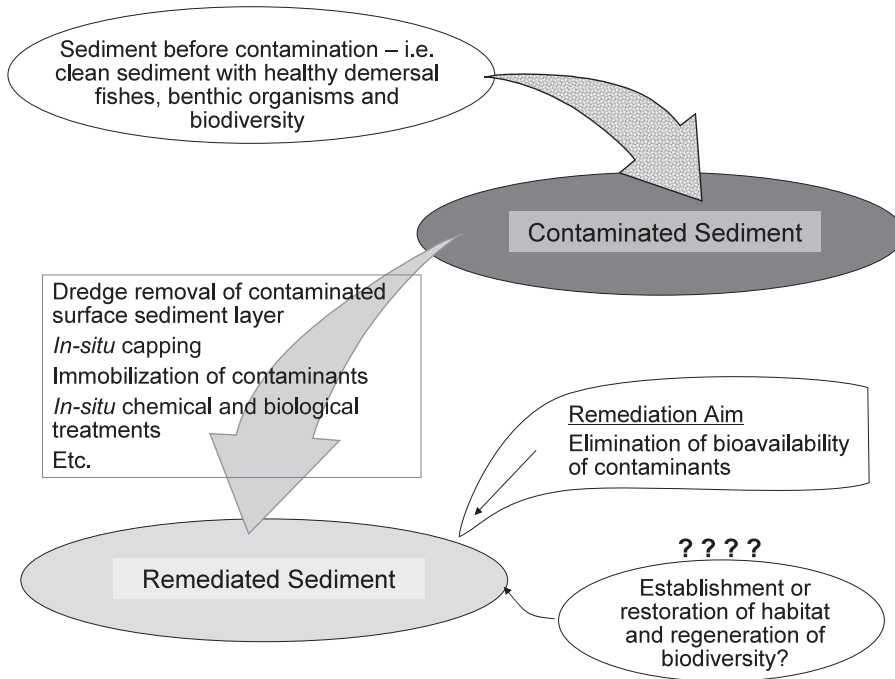


Fig. 2 Two alternative end points for the remediation of contaminated sediments

very pertinent points need to be considered in order to answer this question. First, can the remediated sediment be kept in its freshly remediated harmless or “clean” state over a long term period (presupposing for a moment that it is possible to define when successfully treated sediments can be considered harmless to human health, or “clean”)? And second, is it necessary to consider habitat restoration, species preservation and biodiversity regeneration as part of the remediation technology used to decontaminate sediments, or as a separate and additional set of remediation technologies?

By definition, *sustainability* means the ability to sustain, maintain or preserve. The use of the term *sustainability* in the context of environmental systems refers to the ability of the system to maintain or preserve the initial undisturbed or pre-impact condition, state, or level in the face of assaults on the system. For this to be achieved, the system must be capable of self renewal, self-correction or self regeneration in the face of negative or degradative impacts. The initial undisturbed and pre-impact state of sediments is represented by the top left-hand oval in Figure 2. Here, “sediment before contamination” defines the condition where the uncontaminated sediment is presumed to be both devoid of health-threatening contaminants, and a proper habitat and breeding ground for benthic animals and other microorganisms. It needs to be reiterated that determining the

pre-impact state of a sediment is not easily done, on the grounds that it is difficult both to define what constitutes the pre-impact state, and establish when contamination of the sediment under scrutiny occurred. It is generally assumed that when the contamination of a given sediment is allowed to continue unabated, a concentration of contaminants will be reached where the contaminants threaten human health through bioavailability and transmission through the food chain and impair the breeding grounds and biological diversity of species in the benthic ecosystem.

Sustainability of remediated sediments refers to the ability of the remediated sediments to be preserved in their remediated condition, as befits the specified intent of remediation of contaminated sediments. Anecdotal evidence from present-day application of remediation technology suggests that in most cases, the intent of any remediation scheme is to minimize and/or eliminate health threats to humans and benthic organisms by:

- decontaminating the contaminated sediment via *in-situ* chemical and/or biological means, or removing the contaminated surface sediment layer, or
- implementing immobilization and isolation techniques that render the contaminants in the sediment unavailable for remobilization and resuspension, unable to harm the benthic animals, and unavailable for bioaccumulation or biomagnification.

Any recontamination of remediated sediments will raise threats to human health, and procedures for re-treating re-contaminated remediated sediments are costly and should be unnecessary. The importance of obtaining a sustainable condition for remediated sediments is therefore self-evident. For a remediated sediment to become sustainable, it must: (a) not require re-treatment to preserve its remediated state, and (b) re-establish its original uncontaminated benthic ecosystem. Given the various sources and inputs of contaminants and the various processes contributing to benthic-demersal coupling summarized earlier in Figure 1, it is evident that sustainable remediated sediment preservation may not be easily achieved. A fundamental problem presents itself: can strategies and technologies be developed to facilitate sustainable remediated sediment preservation?

3. Sustainability assessment

Sustainability assessment of remediated sediments is a procedure that is designed to provide knowledge of the extent to which the remediated state of a sediment

can be preserved. The results of any such assessment indicate whether re-contamination of a remediated sediment (a) could occur, therefore requiring amelioration or mitigation, or (b) will reach a level beyond amelioration, therefore necessitating subsequent remediation of the re-contaminated sediment. If habitat restoration and species preservation or biodiversity regeneration form the final set of objectives, it is necessary to specify the corresponding sustainability indicators. In order for sustainability assessment protocols to be performed, knowledge of at least four interacting components must be acquired:

- (i) *Nature and composition of contaminated sediments*: Because of the need to avoid resuspension and remobilization of contaminants from the many forces and fluxes generated in benthic-demersal coupling, it is essential to acquire proper knowledge of how the contaminants are held within the surface sediment layer. This information can also be used to determine the most effective treatment for remediating the contaminated sediment consistent with cost-effective considerations.
- (ii) *Sources and nature of contaminants*: The nature of contaminants together with the nature of the sediment will determine whether resuspension and remobilization of the contaminants is likely to occur. Locating the sources of contaminants provides clues as to the nature and composition of the contaminants. In addition, knowledge of their sources and toxicities to benthic animals and humans is also useful for developing regulations and strategies which manage or control the discharge of contaminants that would eventually find their way into the receiving waters.
- (iii) *Remediation technologies used*: Various strategies for remediating contaminated sediments yield different results with regard to the neutralization and elimination of contaminants in the sediments. The nature of the remediated sediment will have a direct bearing on whether additional measures will be needed to promote sustainability of the remediated sediment.
- (iv) *Requirements for remediated sediment sustainability*: These are dictated by the information obtained from sustainability assessment procedures, the short and long-term human health risks, regulatory attitudes and goals, economics, and site and situation specificities.

The following subsections highlight some of the basic issues, reasons, and requirements for developing detailed knowledge of the four interacting components listed above.

3.1 Contaminants and sediments

The compositional nature of sediments is rarely discussed in detail in the literature dealing with sediments. References to sediment composition are generally in terms of sandy, muddy, silty or granular types of materials. Difficulties in obtaining surface sediment cores reflecting the exact nature of the sediment being sampled have contributed to the lack of detailed compositional description of contaminated sediments. It is often assumed that since the materials constituting most, if not all, sediments originate from land surface soil sources, the composition of sediments will reflect those of the contiguous land mass. A good example of a broad definition is the US EPA (1998) definition of a contaminated sediment as “soil, sand, organic matter, or other minerals that accumulate on the bottom of a water body and contain toxic or hazardous materials at levels that may adversely affect human health or the environment.”

The mobility of contaminants in sediments is a reflection of how well they are attached to the sediment solids — either by forces of interactions resulting in bonding between the contaminants and the solids, or by other means such as chelation or co-precipitation. Since the mechanisms responsible for retention of contaminants in the sediment are functions of both the surface properties of the sediment solids and the contaminants, elaboration of the compositional features of the sediment materials is needed. This will greatly assist in the determination or estimation of the likelihood of resuspension of contaminants.

Whilst the same fundamental mechanisms are responsible for partitioning of contaminants — defined here to mean the physical and chemical mass transfer of contaminants from the water to the surfaces of the sediment solids — and the bioavailability of contaminants in freshwater and marine sediments, considerable differences exist between freshwater and marine sediments in respect to how these mechanisms react in the two different water environments. The processes of partitioning are sensitive to pH changes and the presence of ligands in the interstitial water. The range of pH in seawater is relatively small compared to that of freshwater in rivers and lakes because the chemical composition of seawater is not as variable as that of freshwater in lakes and rivers. In the latter case, the chemistry of the freshwater is a direct reflection of the chemistry of the run-offs from the land mass surrounding the freshwater body, and the chemistry of the groundwaters feeding the receiving freshwaters. Studies on the effect of salinity on the bioavailability of trace metals (McLusky et al. 1986) show that in general, the toxicity of the metals varies inversely with the salinity of the host water. However, as shown by Voyer and McGovern (1991), differences in osmoregulation of metal uptake between different marine species make it difficult to generalize upon the relation between toxicity and salinity. In respect

to the effects of salinity on the availability of organic chemical compounds for uptake, studies reported by Geyer et al. (1982) and Veith et al. (1979) indicate no significant differences for bay mussels and fathead minnows respectively. The relationship between the octanol-water partition coefficients and the bioconcentration factor were found to be applicable for both environments.

More recent direct evidence of sediment depositional compositional features reflecting the surface soil characteristics of contiguous regions has been reported by Quigley et al. (1985). Tests were performed on recovered samples from soft, freshwater varved clays from the proglacial Lake Ojibway at Matagami in Québec Province in Canada. The clays (former sediments of the lake) were deposited over a period of just over 600 years during the two advances and retreats of the Hudson glacier. The results of tests show two sources of soils for the varved clay sediments: the first, carbonate-deficient Precambrian crystalline igneous rocks to the east of the proglacial lake, and the second, carbonate-rich Paleozoic rocks from the lowlands to the west of the lake. Analyses of the composition of the various varves showed clay minerals (illite, chlorite and some smectite) together with quartz, feldspar, calcite, dolomite and amphibole — all reflective of the minerals in the Precambrian and Paleozoic rocks to the east and west of the lake. Many more examples of studies on cores retrieved from former lake beds and marine incursions can be cited to show how the composition of sediments is a reflection of the nature of the soil-source materials in the surrounding or adjacent land mass. A geochemical study of sediment cores allows one to obtain a record of natural and, if appropriate, human events impacting on the land mass defining the source region and/or drainage basin for the sediments.

Studies of the surficial bottom sediments of contained water bodies such as lakes provide direct evidence of sediment compositional relationships to depositional environment. One such study reported by Nelson and Lister (1995) found strong interrelationships between recent depositional lake environments and surficial sediment deposits — with source material from the catchments to the south of the lake. Solids in the sediments such as clay minerals, oxides and hydrous oxides, carbonates, sulphides, and soil organic matter act as major sinks and reservoirs for many kinds of contaminants. An earlier study by Dossis and Warren (1980) on the near-shore marine sediments near a large lead smelter in Spencer Gulf, South Australia, showed the presence and bonding of zinc, lead, and cadmium in the sediments with the organic debris, conglomerate particles, feldspar, magnesium calcite, mica, kaolin minerals, and pyrite (amongst other types of primary minerals and oxides) in the sediment. Results of analyses of sediments recovered near shipyards in San Diego Bay in California showed them

to be heavily contaminated with heavy metals, with concentrations up to 5500 ppm for copper (Meiggs 1980). Interestingly, the greatest retention of the heavy metals occurred in the uppermost portion of the sediment layer. This testified to the sorption capacity of the soil-material-sinks in the sediment.

More recently, considerable evidence of widespread contamination by organic chemicals and inorganic chemicals including heavy metals in fresh-water and marine sediments can be found in the literature. Contaminants widely reported in sediments consist of nutrients such as phosphorous and nitrogen compounds (e. g. ammonia), persistent organic pollutants (e. g. PCBs), hydrocarbons, petroleum products, oil and grease, and heavy metals such as mercury, lead, manganese, cadmium, selenium, zinc, etc. The US EPA's comprehensive report on the severity of contamination of sediments in surface waters of the United States (US EPA 2004) is a good example of the information available. In all the accounts available, the sources of contamination are attributed to their specific point and non-point sources. Fulweiler and Nixon (2005) report on the export of nitrogen, phosphorus, and suspended solids from the Pawcatuck River watershed in New England to Little Narragansett Bay. Measurements of nitrogen showed dissolved inorganic nitrogen and phosphorous and organic forms of nitrogen and phosphorous. The inorganic forms were found to be at least twice the proportion of organic forms. Yong (1995) and Yong et al. (1995) have reported on the presence of a large group of heavy metals (from chromium to zinc), "oil and grease" and PCB in sediments retrieved from Lachine bay and the Lachine canal, immediately south-east of the island of Montreal in Canada. The sources for these contaminants were chemical and metals industries located upstream from the contaminated sediments.

Sediments recovered in many parts of the Great Lakes in the United States, reported to consist of particles of rock, soil and decomposing wood and shell, show the presence of land source and deposited airborne pollutants, plus chemicals from sources as far away as hundreds and thousands of miles (US EPA Great Lakes National Program Office 1994). Pollutants reported as present in the contaminated sediments consisted of heavy metals and toxic organic chemicals from both municipal and industrial wastes, as well as herbicides and pesticides from farm runoff.

Most of these pollutants are, in one fashion or another, attached to soil organic matter (SOM) and/or mineral particles when they enter into the aquatic regime. Chlorinated hydrocarbons and other hydrophilic compounds for example are commonly found with SOM. On the other hand, it is not unusual to find the lower water-solubility compounds such as PAHs and PCBs. bonded to the particulate minerals (primary and secondary minerals). The heavy metals are

generally found in association with clay minerals because of physical and chemical sorption bonding. They can also be found as co-precipitates with several oxides, sequestered in organic matter. Resuspension and remobilization of the contaminants in sediments result from bioturbation, benthic boundary layer flow, chemical reactions and exchange, and other mixing forces that contribute directly to benthic-demersal coupling. Benthic animals and microorganisms are significant participants in the metabolism of petroleum products in the sediments. The result of activities associated with these organisms can be seen in terms of physical mixing and increased water solubilization and resuspension of the chemicals.

It bears repeating that the complex nature and composition of sediments makes it very difficult to properly quantify and, in particular, specify the transport and fate of pollutants found in the sediments. One way to understand the various forces and bonds established between the various sediments constituents and pollutants is to refer to knowledge of the interacting mechanisms between soil fractions and pollutants, as well as studies of the abiotic and biotic reactions in their interacting systems.

3.2 Remediation objectives and technology

The principal objective in remediating contaminated sediments is to make the sediments harmless in terms of their threat to ecological and human health. Recognizing that the primary threat to human health is the bioaccumulation and biomagnification of contaminants reduces the problem to one of restricting the bioavailability of the contaminants. This requires the reduction or elimination of the toxicity of the contaminants and removal of the potential for their resuspension and remobilization.

With the exception of general reductions in the concentration of contaminants as a means for toxicity reduction or elimination, the methods or paths for toxicity reduction differ for inorganic and organic contaminants. For example, in the case of heavy metals (the primary inorganic toxic contaminants), toxicity reduction depends upon the elimination of the availability of the metals. This is best accomplished by immobilizing them by the use of chemical or physical means. For organic chemical compounds, biological processes are popularly used to (a) transform these kinds of compounds to intermediates that are less toxic (biotransformation), and (b) break down these compounds by enzymatic and other degradative actions to intermediates or end products that are less toxic (biodegradation). Some of these processes are described in Mulligan and Yong (2006), in relation to the natural attenuation processes inherent in many soil-composed types of sediment.

Figure 3 shows that resuspension and remobilization of contaminants in the sediment take place primarily in the top portion of the contaminated surface sediment layer. Bioturbation and benthic boundary layer flow, including tidal exchange, in general affect only the top 30 centimetres of the surface sediment layer. Boyer et al. (1990) have reported on a disturbance layer of about 30 centimetres for Burbot fish in freshwater sediments and corresponding depths of surface sediment layer disturbance from burrowing by fiddler crabs have been reported by Warner (1977) in marine sediments. In order to distinguish between this disturbed layer and the remaining underlying contaminated layer, the top layer can be called the “turbulent layer”. The contaminants in this turbulent layer are subject to resuspension and remobilization, and for this reason pose a threat to benthic animals living in the demersal zone. The contaminants below the turbulent layer are likely to remain contained in the sediment, except in the case of violent wave action which can physically disturb contaminants in the bottom portion of the surface layer to the point at which they become resuspended. Note however that continuous chemical and biological activities in this lower portion of the contaminated surface sediment layer will result in exchanges with the overlying sediment layer.

Various procedures currently popular for use in remediating contaminated sediments fall into four main categories. They include 1) the removal of the contaminated surface sediment layer with or without replacement of fresh uncontaminated material, 2) *in-situ* capping of the contaminated surface sediment with an impervious or semi-pervious cap system consisting of a

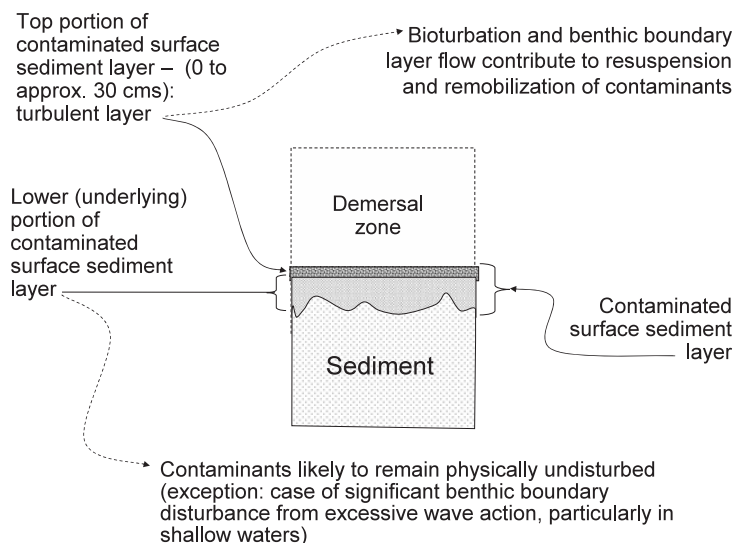


Fig. 3 Schematic illustration of contaminated surface sediment layer

synthetic membrane covered by clean coarse-to-fine granular material and rock, 3) isolation, immobilization and containment of the contaminated sediments, and 4) *in-situ* chemical and biological treatments. All four can be seen summarized in Figure 4. Here, (A) shows the initial condition of a contaminated surface sediment layer on top of the underlying uncontaminated sediment and (B) represents the removal of the contaminated surface sediment layer as a remediation option. The end product of the removal strategy is always a fresh uncontaminated sediment surface: whether this is achieved simply by removal or by the replacement of the removed contaminated surface sediment layer with fresh uncontaminated material is a function of the remediation strategy employed. Treatment of the removed contaminated sediment material *ex-situ* is a requirement for disposal of the material on land. Whilst it is not common to re-use the *ex-situ* treated material as replacement for the surface sediment layer, anecdotal evidence suggests that this sometimes takes place.

Capping, immobilization, isolation and containment are examples of physical sediment surface barriers to re-infection or re-contamination of the surface sediment layer. These are represented by (C), where an *in-situ* cap consisting of a membrane is shown placed on top of the contaminated sediment. Crushed stone and gravel are frequently used to cover this membrane. Since no attempt is made in this case to decontaminate the contaminated sediment, the physical capping restraints serve as the sole agents for prevention of resuspension and remobilization of the contaminants in the sediments. In essence, the *in-situ* cap

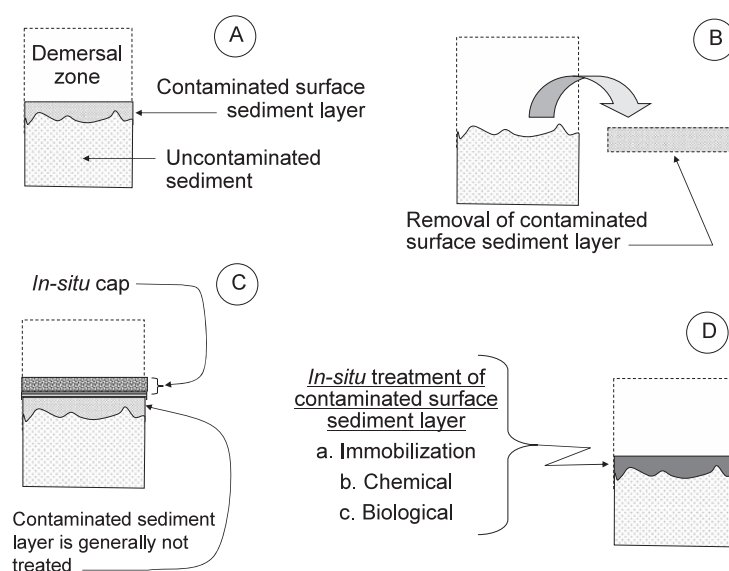


Fig. 4 Various remediation procedures and strategies

prevents the formation of the turbulent layer, in this way reducing or even eliminating bioaccessibility. A good example of containment remediation of contaminated sediment has been reported by Hosokawa (1993). It took place in Minamata Bay on the island of Kyushu in southern Japan, where isolation of 58 hectares of the bay area showing the highest levels of mercury-contaminated sediments was achieved by the use of cofferdams. The cofferdam containment area accepted in addition more than one million cubic metres of contaminated sediments brought in from areas outside the containment area. The contaminated sediments in the containment area were capped with volcanic ash, sand, and geotextiles. Since the capped containment area was below ground level (grade), extra fill material has since been brought in to bring the level of the remediated containment area to ground level — i. e. the area has been filled and brought to grade.

(D) in Figure 4 shows three final treatment options. The first, immobilization of the contaminated sediment layer, is obtained through the use of fixing agents such as cement and other kinds of hardening grouts and bituminous materials. The end purpose of this technique is to completely immobilize any contaminants which have settled in the matrix of the “fixed” sediment layer. The two remaining options shown in (D) refer to *in-situ* injections of chemical or biological aids as treatment procedures. *In-situ* chemical or biological treatments of contaminated sediments seek to establish biotic and/or abiotic chemical reactions amongst the contaminants in the sediments. The ultimate goal of these treatments is to reduce or eliminate the toxicity threat of the contaminants resident in the sediment. This is achieved by two principal mechanisms: the creation of more robust bonding between the contaminants and the solids in the sediments and the biotransformation of organic chemical pollutants to lesser or non toxic intermediates.

It is important to note that although sustainability requirements are not necessarily implicit in the structuring of the contaminated sediment remediation procedures described above, all these procedures share the same objective in seeking to deny the resuspension and remobilization of the contaminants in the sediment. More recently, with a view to sustainability, the assimilative capacity of sediment materials has been increasingly exploited as a means to attenuate the contaminants in the contaminated sediments. The term *natural recovery* (NR) has been used to identify the results of contamination attenuation in the sediments through natural processes. The processes involved are in almost all respects similar to those available in the *natural attenuation* (NA) treatment processes used in the solid land environment, described in Yong and Mulligan (2004). The primary processes involved in NR fall under the category of bioremediation or

biotransformation. These complex processes — conditioned not only by the natural microbial communities and metabolic processes, but also by the nature of the organic compounds and the other sediment constituents — are discussed in more detail in section 5.1 of this paper.

4. Strategy for remediated sediment sustainability

It is commonly assumed that freshly remediated sediments are “clean”, or in other words, that they no longer pose indirect or direct threats to human health having been denied the opportunity for bioavailability and bioaccumulation by the remediation treatment. As should be clear from the previous discussion on sustainability objectives, the sustainability goal for a remediated sediment is a 100 per cent self-capability to preserve its “clean” state in the long term. Three conditions are essential if this goal is to be achieved. First, the treatment technology used to remediate the originally contaminated sediment must be one that can maintain the remediated sediment in its “clean” state; second, voluntary and regulatory controls should be used to limit or even eliminate the further input of contaminants into the demersal zone and the benthic environment; and third, all suspended solids in the demersal and pelagic zones associated with the remediated sediment should be removed. Figure 5 illustrates a five-part strategy

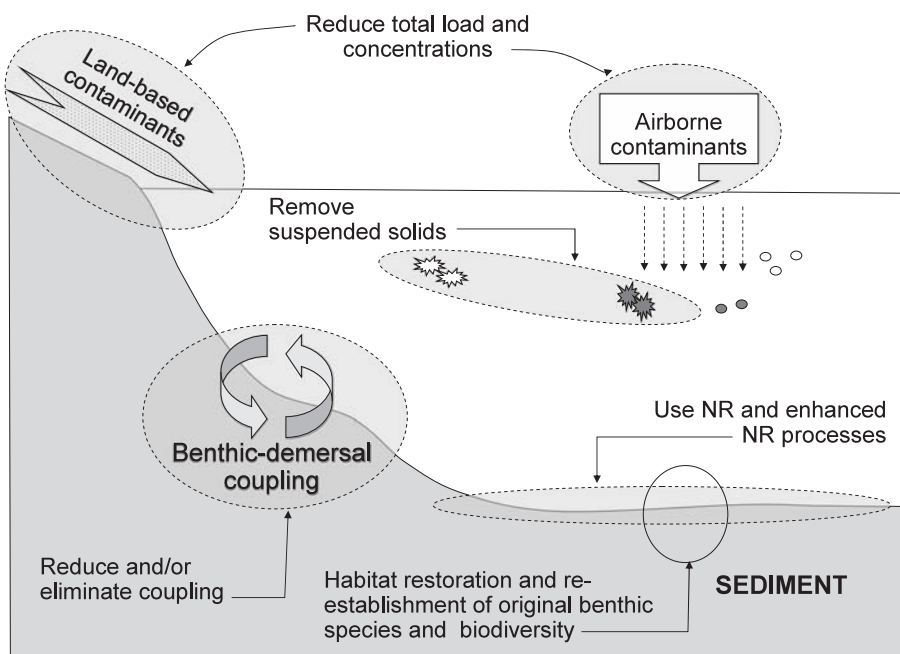


Fig. 5 A five-part strategy to obtain complete sustainability of the remediated sediment

by which it becomes possible to obtain the complete sustainability of a remediated sediment.

The basic elements of the strategy include:

- (i) Reduction or elimination of land-based contaminants discharged into the receiving waters and noxious airborne gases and particulates, via a combination of voluntary action and regulatory controls.
- (ii) Reduction of suspended solids in the demersal zone above the affected sediment by “cleaning-up” the water in the affected region (provided that land-based and airborne contaminants and noxious substances are reduced, the water clean-up should be a one-time action).
- (iii) Reduction or elimination of the negative impacts from benthic-demersal coupling. This is a direct function of the treatment used to implement the remediation strategy.
- (iv) Exploitation and enhancement of natural recovery (NR) processes. These processes are inherent properties of the sediment and can be fully exploited and even enhanced if proper knowledge and understanding of the material and system parameters are obtained.
- (v) Restoration of habitat and re-establishment and/or regeneration of benthic species and biodiversity. The normal course of action would involve the provision of necessary nutrients and a basic stock of benthic animals and microorganisms from regions sharing similar characteristics to the remediated sediment zone.

In an ideal situation, all the necessary actions outlined in the five-part strategy can be well implemented by industry with the participation of specialists in benthic ecosystems and biodiversity. Whether industry will carry out all these actions voluntarily or in the absence of regulatory controls remains an open question, however. The “real world” situation recognizes that there will always be some measure of contaminants entering the receiving waters from land and airborne sources. Accordingly, the prudent course of action is to ensure that the sustainability measures listed in the five-part strategy are given serious consideration in the short-term, with a view to their implementation in the long-term.

5. Measures for sustainability management

The elements that are manageable within the present context of *sustainability management* of the remediated sediments are those elements identified in (i)

through (iii) in the five-part strategy laid out in the previous section. A fundamental requirement for sustainability is the capacity for self-renewal or self-regeneration without human interference. In the case of remediated sediments, this prompts the use of natural recovery processes to neutralize, decontaminate, and/or render any incoming contaminants unavailable to benthic animals.

5.1 Natural recovery as a fundamental building block

Natural recovery or NR processes, so-named because of the ability of natural properties and characteristics of the sediment material to “clean itself” through self-remediation and self-correcting processes, can be considered the basic building block for making remediated sediments sustainable. The key to NR success lies in the reactive compatibility between the contaminants and the composition of the sediment. Many of the basic processes involved in NR are similar to processes involved in the natural attenuation (NA) of contaminants in soils. Figure 6 shows the principal mechanisms and processes involved in NR. Not all of these are common to all sediments, as wide variation in the composition of sediments and the dynamics of their immediate benthic-demersal environments affect the processes by which they naturally recover. Three basic

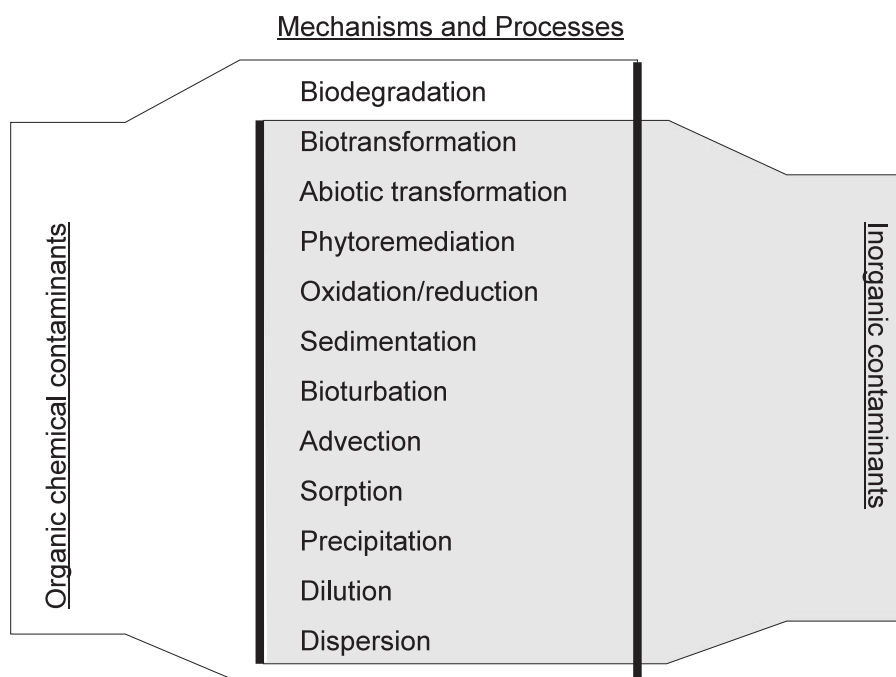


Fig. 6 Major processes involved in reactions between organic chemical and inorganic contaminants in sediments

groups of mechanisms and processes nevertheless emerge, which may be summarized as follows:

- (a) Physical: sedimentation, bioturbation, advection, dispersion, dilution, diffusion, sorption.
- (b) Biological: bioremediation, biotransformation, biodegradation,
- (c) Chemical: oxidation-reduction, chemisorption, transformation, precipitation.

Assessment of reactions between contaminants and sediment materials can be grouped under two classes: the first dealing with inorganic contaminants, and the second, with organic chemical contaminants. The major proportion of reaction processes between inorganic contaminants and sediment fractions are physical and chemical. For heavy metals, pH is an important factor in their bioavailability. Most of the metals are associated (sorbed or “bound”) with carbonates, iron and manganese oxides, and in some instances, in the inner lattice structure of the clay minerals. Typically, zinc and lead tend to be bound with the carbonates that exist as solids in the sediment, or they may even exist as exchangeable cations. Copper on the other hand would most likely be found in association with the oxides in the sediment, and chromium and nickel would most likely be bound with the lattice structure. A great deal depends on the proportions of the various heavy metals during discharge and initial contact with the sediment solids. Competition for sorption sites and bonding mechanisms between the various heavy metals will determine how they are partitioned between the various sediment materials.

The ability for sediments to accumulate more heavy metal contaminants during NR depends not only on accessibility and availability to sorption-bonding sites for the different sediment fractions, but also on whether these sorption sites have been exhausted. Luoma (1989) has shown that in oxic sediments, the availability of heavy metals for bioaccumulation is a function of their ability to bind onto organic-carbon and sorbed by the oxides of iron and manganese. This has been confirmed by Yu et al. (2001). Bioturbation and bioirrigation can physically disturb sediments. These actions may serve to release metals in the sediments. In anoxic sediments, these disturbances provoke the release of metals associated with acid volatile sulphides through oxidation of the sediments.

By contrast, biological processes play a major role in the NR of sediments contaminated by organic chemicals. They can transform organic chemical contaminants via biotransformation to intermediates that are less toxic or even non-toxic. The breakdown of organic chemicals and other carbon containing

materials by enzymatic and other degradative actions provide the nutrients and carbon for development of microbial populations in the sediment. In time, the target organic chemicals in the contaminated sediments are converted or transformed to intermediates or end products that are less toxic or completely harmless. This process, known also as biodegradation, is described in detail in Mulligan and Yong (2004). Because of the many complex processes involved in both biotransformation and biodegradation, the biological treatment of organic chemical contaminants in sediment requires knowledge of the natural microbial communities present in the sediment in terms of their structure, function and metabolic processes. As these processes are generally organic chemical specific, their study requires the thorough examination of individual chemical compounds, generally through bench-scale experiments. Nevertheless, biological treatment of sediments is a powerful tool in the NR repertoire and can be well exploited. Biological treatments work particularly well in cases where environmental circumstances permit acceleration of biological processes through enhancement techniques such as the addition of nutrients and other growth substrates, together with electron donors and acceptors to promote greater microbial activities (biostimulation), or the addition of other supportive microorganisms to indigenous microorganisms to increase the effectiveness of the biological treatment (bioaugmentation).

Determining whether NR will be applicable as a remediation or sustainability tool requires the assessment of (a) the natural attenuation properties of the sediments, (b) the nature and distribution (including concentrations) of the various kinds of contaminants, (c) the previous physical and chemical interactions with contaminants in the sediment, (d) intrinsic or natural bioremediation history and capability, and (e) other factors associated with the transport and fate of contaminants. The level of detail and information required is site specific. Knowledge of all these factors can be compiled to support an investigation generally called *NR Lines of Evidence*. Although similar in some aspects to lines of evidence rationale used to monitor natural attenuation as a remediation tool on solid land environment, the activities and requirements to establish *NR Lines of Evidence* differ markedly from activities used to monitor the natural attenuation of soils, largely because of the benthic-demersal coupling phenomenon (shown for example in Figure 1) and the dynamic nature of sediments. In establishing an *NR Line of Evidence*, there are essentially four characterization requirements with corresponding sets of activities that must be established *en route* to the final objective — i. e. to determine the capability and sustainability of NR for a given sediment.

The four characterization requirements together with their activities are

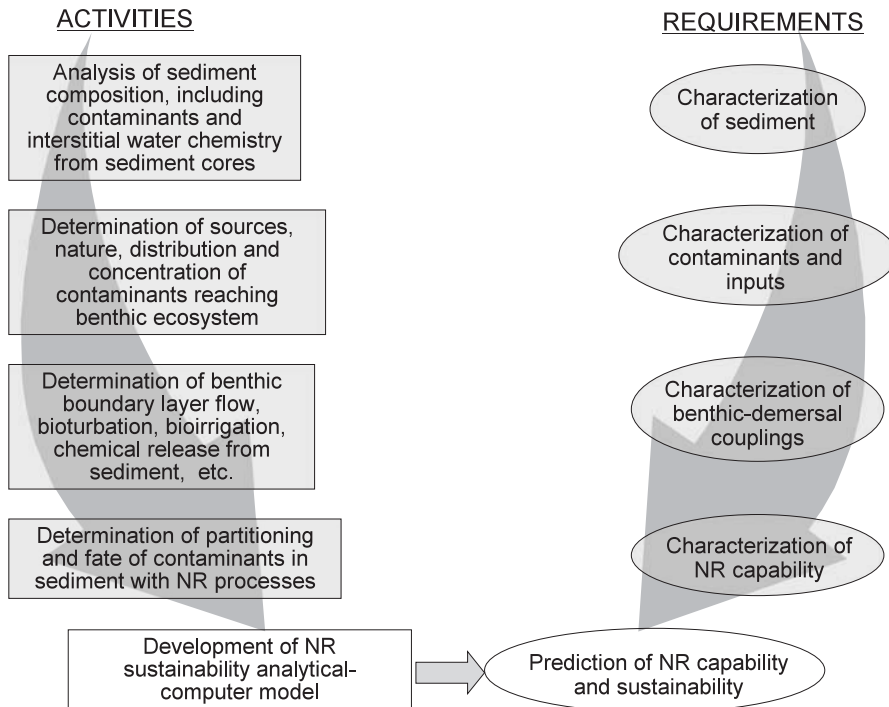


Fig. 7 Activities and requirements in support of NR Lines of Evidence

shown in Figure 7 and are summarized as follows:

- **Characterization of sediment:** Notwithstanding the dynamic nature of the sediment, a detailed understanding of the composition of the materials that comprise the sediment is essential. Information relating to the surface forces of the sediment solids and organic debris is likewise important, since these interact directly with the contaminants in the sediment. Other vital information relates to the pH, redox potential and chemistry of the interstitial water, and cation exchange capacity and specific surface area of the sediment solids.
- **Characterization of contaminants and inputs:** Knowledge of the specific source(s) of contaminants yields information as to their types and properties. Information as to how they are delivered to the sediments is also vital, with a view to controlling and regulating future contaminations.
- **Characterization of benthic-demersal couplings:** These couplings are mainly a result of disruptive mechanisms and processes caused by the actions of benthic animals and interface wave actions. Benthic environ-

mental forces can detach attached contaminants from sediment solids, leading to the resuspension and remobilization of contaminants. For this reason, their study is vital.

- Characterization of NR capability: This includes knowledge of all the partitioning mechanisms and bonding capacities established between contaminants and sediment solids. Knowledge of these capacities can be obtained from laboratory partitioning tests and sequential selective extraction procedures on retrieved samples. Since evidence of intrinsic remediation is one of the central issues, identification of microorganisms is a prime requisite.

The results from the four activities can be used as inputs to develop an analytical-computer model for the assessment and prediction of NR capability and sustainability. Initial attempts to predict NR capability have used fate and transport models developed for use in predicting land subsurface contamination. These have had questionable success, partly because of the lack of validation information and partly because of the limitations of the land fate and transport models.

5.2 Enhanced natural recovery (ENR)

Enhancement of natural recovery processes provides the remediated sediment with greater capability to maintain its “clean” freshly remediated status. Chemical and biological aids are commonly used as enhancement tools. Chemical treatments alter the immediate environmental conditions to allow for the development of microbial populations that can degrade the organic chemical contaminants in the sediment. Biological treatments can take the form of bioaugmentation and biostimulation — both of which increase the microorganisms’ genetic capability to use the organic chemical contaminants as sources of carbon and energy. These chemical and biological techniques are designed to help the microorganisms already present in the sediment hasten the biodegradation processes.

5.3 Soil-sediment material as a design NR remediation tool

With the proper choice of soil-sediment fractions (constituents) it is possible to “design” a soil-sediment that can provide optimum attenuation of contaminants. This greatly increases the sustainability of NR as a remediation tool, so long as the rate of recovery or attenuation is equal to or higher than the rate of sediment contamination. This capability makes “designer” soil-sediments an excellent choice for replacing surface sediment material in situations where dredged or

removed sediments require replacement, as for example in the case where new habitats need to be established for benthic organisms.

6. Sustainable remediation

6.1 Water column purification

Technologies for remediating contaminated sediments can be designed to produce remediated sediments that remain “freshly remediated” in the long term. Their success in terms of sustainability depends, however, on the successful implementation of contaminant source control and elimination of contaminant input into the receiving waters and underlying sediments. In contained or semi-contained water bodies such as lakes and bays, “clean-up” of the water column over the remediated sediment aids sustainability requirements. The demonstration vessel test facility currently being operated in Kasaoka Bay, Japan (Fukue et al. 2004) provides a good example of this: the vessel uses filtration techniques to purify contaminated seawater and monitor the water for suspended solids, chemical oxygen demand, pH, etc.

6.2 NR-based sustainable solutions

Prudent remediation technology recognizes that source control and management of land-based pollutants will not fully eliminate their discharge into the receiving waters. For this reason, sustainable remediation strategies should take NR as their central precept and produce remediated sediments that are self-cleaning. Figure 8 shows how this might be done. In the figure, (A) shows a contaminated sediment layer overlying uncontaminated sediment and (B) shows a conventional procedure involving removal of the contaminated surface sediment layer. A choice arises at this juncture: to leave the situation as it is represented in (B), or to replace the removed layer with clean sediment material. The “no replacement layer” option is premised on the assumption that the remaining uncontaminated sediment is capable of eventually (and naturally?) providing a regenerated habitat for benthic organisms. In this case, for sustainable remediation to be successful, the remaining uncontaminated sediment must be capable of self-cleaning. In the absence of human intervention, this means that the uncontaminated sediment must possess NR capabilities that can accommodate any incoming contaminants.

A third option is shown in (C), where biotreatment is used to enhance the NR capability of the uncontaminated sediment. If a surface sediment layer replacement is to be used, as in (D), it is possible to biologically treat the uncontaminated sediment first, before the placement of an NR-capable

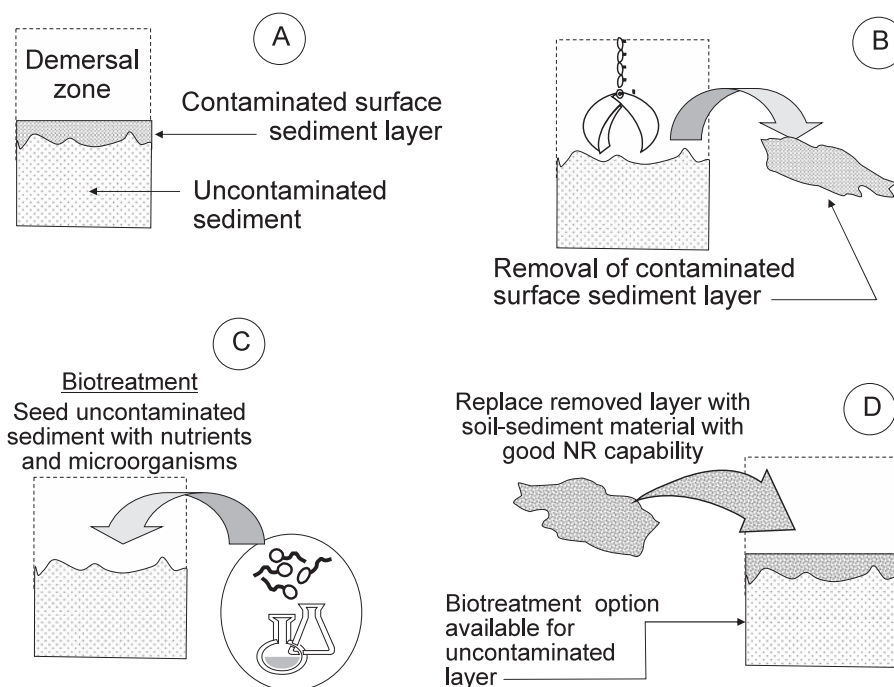


Fig. 8 Use of NR as primary component for sustainable remediation of contaminated sediments

replacement layer. The same biotreatment technique can be used to increase the NR capability of the replacement sediment layer. Under normal circumstances, soil materials with capabilities for sorption of inorganic and some organic contaminants are chosen for their high NR capacities. Their role as NR materials can be further enhanced with the addition of microorganisms and nutrients.

In-situ capping can also be made sustainable as a remediation option, as shown in Figure 9. In this case, the choice can be taken to treat, “fix” or immobilize the contaminated surface sediment layer — as in (B) — before placing the *in-situ* cap over the untreated or treated contaminated sediment layer. Introducing a competent NR soil-sediment layer on top of the cap, as in (C), provides not only a more favourable habitat restoration potential, but also a more sustainable habitat, owing to its “self-cleaning” capability.

Other methods exist for remediating contaminated sediments, but few are sustainable without some form of human intervention or source control on pollutant discharges. NR with its various enhancements is the key to a sustainable remediation programme. For it to be fully successful, however, a competent monitoring programme needs to be structured to determine whether the various processes designed to attenuate the pollutants and contaminants are functioning at a level that meets expectations.

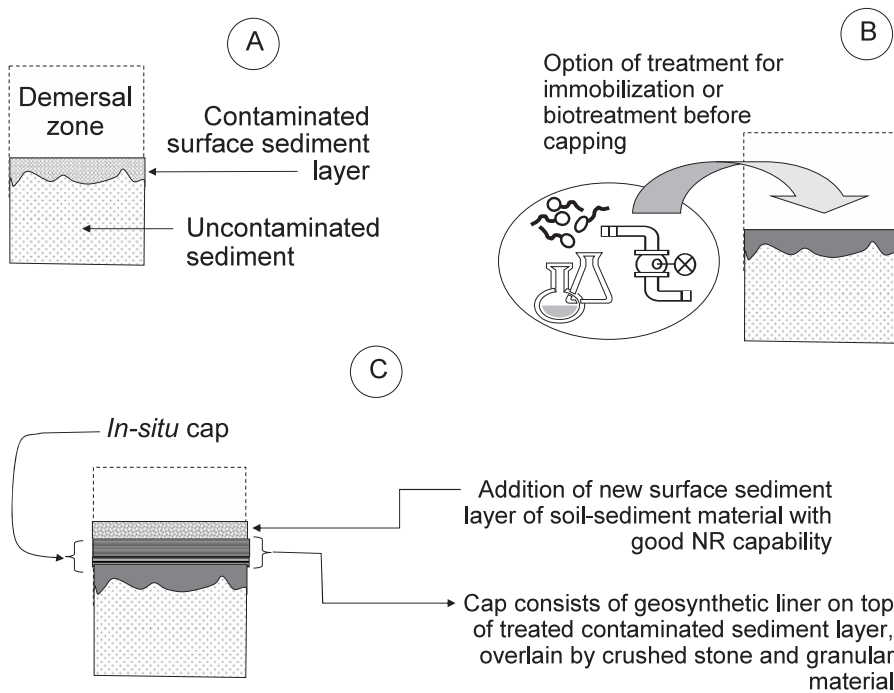


Fig. 9 Use of NR-capable soil-sediment on top of cap to promote sustainable remediation

7. Sustainability indicators

7.1 Determination of sustainability indicators

Sustainability indicators used for monitoring the remediated state of contaminated sediments previously subjected to some form of remediation can be very simple or complex. Much depends on the choice of specific markers or targets and the level of detail required to establish sustainability. If pre-contamination or “clean” sediments can be found in the same region, they can provide baseline information which can be used to specify the indicators. Some typical indicators might be (a) the level of bioturbation and bioirrigation, (b) the distribution of partitioned contaminants, (c) the nature and concentration of contaminants in the interstitial water, and (d) biological diversity. Figure 10's summary sketch of the protocols leading to sustainability assessment of a remediated sediment is based primarily on the distribution and concentrations of contaminants in the remediated sediment.

It is useful to note that there are several criteria that can be employed to declare when sustainability of the remediated sediment has been achieved. In the case of the protocols shown in Figure 10, the presence of contaminants in the

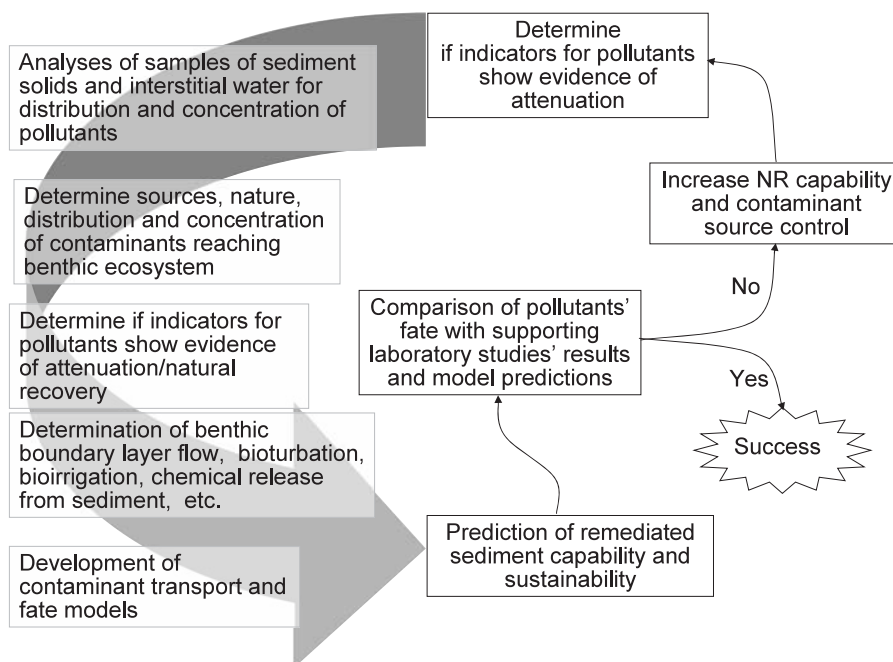


Fig. 10 Required activities for sustainability assessment of remediated sediment based on distribution and concentration of contaminants in remediated sediment

remediated sediment is the yardstick by which sustainability will be assessed. The indicators listed in the first decision box (“Determine if indicators for pollutants show evidence of attenuation”) refer to distributions and concentrations of target species of contaminants in the surface sediment layer. This is equally applicable to the NR layer lying on top of the *in-situ* cap in Figure 9, the NR layer in option (D) in Figure 8, or the treated layers in options (B) and (C) in Figure 8.

The activities necessary to satisfy the requirements for determining indicators are shown on the left-hand side of the illustration in Figure 10. These include:

- Procurement of sediment and interstitial water samples for analyses to determine the nature and distribution of contaminants attached to sediment solids and present in the interstitial water.
- Determination of the sources of contaminants finding their way into the receiving waters and into the remediated sediment. An accounting of the suspended solids in the water column above the remediated sediment is required. The results obtained can be used to inform decisions regarding the implementation of water-column clean-up procedures. In the case that these procedures are already being implemented, the activities will allow

assessors to determine their efficacy.

- Associated laboratory tests and studies for partitioning the kinds of contaminants found in the sediment, together with studies on the intermediate products of typical organic chemical pollutants found in the sediment.
- Assessment of the potential for bioaccessibility, by determining the various physical and chemical forces that will result in resuspension and remobilization of contaminants in the turbulent layer.
- Development and implementation of analytical-computer models developed to predict or analyze the transport (of contaminants) and fate of contaminants in such an environment.

Some would argue that the required activities identified in Figure 10 are considerable and unnecessarily detailed. Much depends on the vulnerability of the remediated sediments to re-infection and the degree of threat posed by re-infected sediment to human health and the benthic ecosystem. Absent from the schematic shown in Figure 10 is a decision-box dealing with risk assessment and management. The risk of re-infection of the remediated sediment is a central issue in sustainability assessment and deserves full attention and elaboration in a separate discussion on the subject.

7.2 Habitat restoration, species re-establishment and biodiversity regeneration

It is recognized that considerable effort in the development of current technologies for remediating contaminated sediments is directed towards contaminant-removal, contaminant-isolation, or reduction in the toxicity of contaminants. With good reason, remediation treatment goals focus predominantly on contaminant removal or rendering the contaminants in sediments harmless. Treatment technologies that seek also to restore habitat and re-generate biodiversity in the benthic ecosystem have not received comparable attention. It is generally thought within remediation treatment circles that “nature will take care of itself” once a clean benthic ecosystem is obtained. This may indeed be true: the question arises, however, as to how long a time-scale is necessary for this kind of natural regeneration to occur.

7.3 Ultimate sustainability indicators

Restoration of habitat and re-establishment of biodiversity are the ultimate sustainability indicators for remediated sediments. Specification of sustainability indicators requires the acquisition of clean-sediment baseline information for the purpose of comparison. Protocols for establishing when ultimate sustainability is

attained require exhaustive sampling activities and analyses of species, colonies and distribution of benthic organisms. The nutrients and other food sources for the organisms also need to be taken into account in establishing sustainability indicators, as do factors such as species diversity, natural communities and other related biomarkers, all of which are important aspects of ultimate sustainability.

8. Concluding remarks

Remediated sediments are not necessarily remediated to the extent that all their contaminants have been removed, nor are they always “clean” in terms of being devoid of contaminants. Indeed, on the basis of remediation technologies currently applied (with the exception of technologies which physically remove the entire contaminated sediment layer), it must be concluded that no remediated sediments are truly “clean”: they are simply remediated to the extent that the threats posed by the contaminants within them to human and sometimes benthic health are neutralized or eliminated. The presence of contaminants in remediated sediments notwithstanding, successful application of the technologies currently in use deny bioaccessibility and bioavailability by reducing the toxicity of the contaminants and impeding their resuspension and remobilization.

The objective of sustainable remediated sediment preservation requires action on three fronts: 1) source control of contaminants entering the benthic ecosystem, 2) implementation of a “self-cleaning” remediated surface sediment layer that maintains the freshly remediated state of the sediment, and 3) restoration of habitat and re-establishment of biodiversity. Action on the first two fronts only will result in a “clean” but ultimately sterile sediment bed environment. Steps therefore need to be taken to ensure that the proper setting is established for habitat restoration to take place. Human intervention in providing the necessary elements for restoration of habitat and re-establishment of biodiversity, after or during remediation of the contaminated sediment, will provide for sustainable remediated sediment preservation.

Acknowledgements

The author expresses deep appreciation to the reviewers of his original manuscript for their constructive suggestions and remarks. These, together with the editor’s valuable input and comments, have been incorporated in the revisions to this paper.

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